PICOSECOND LASER PULSES

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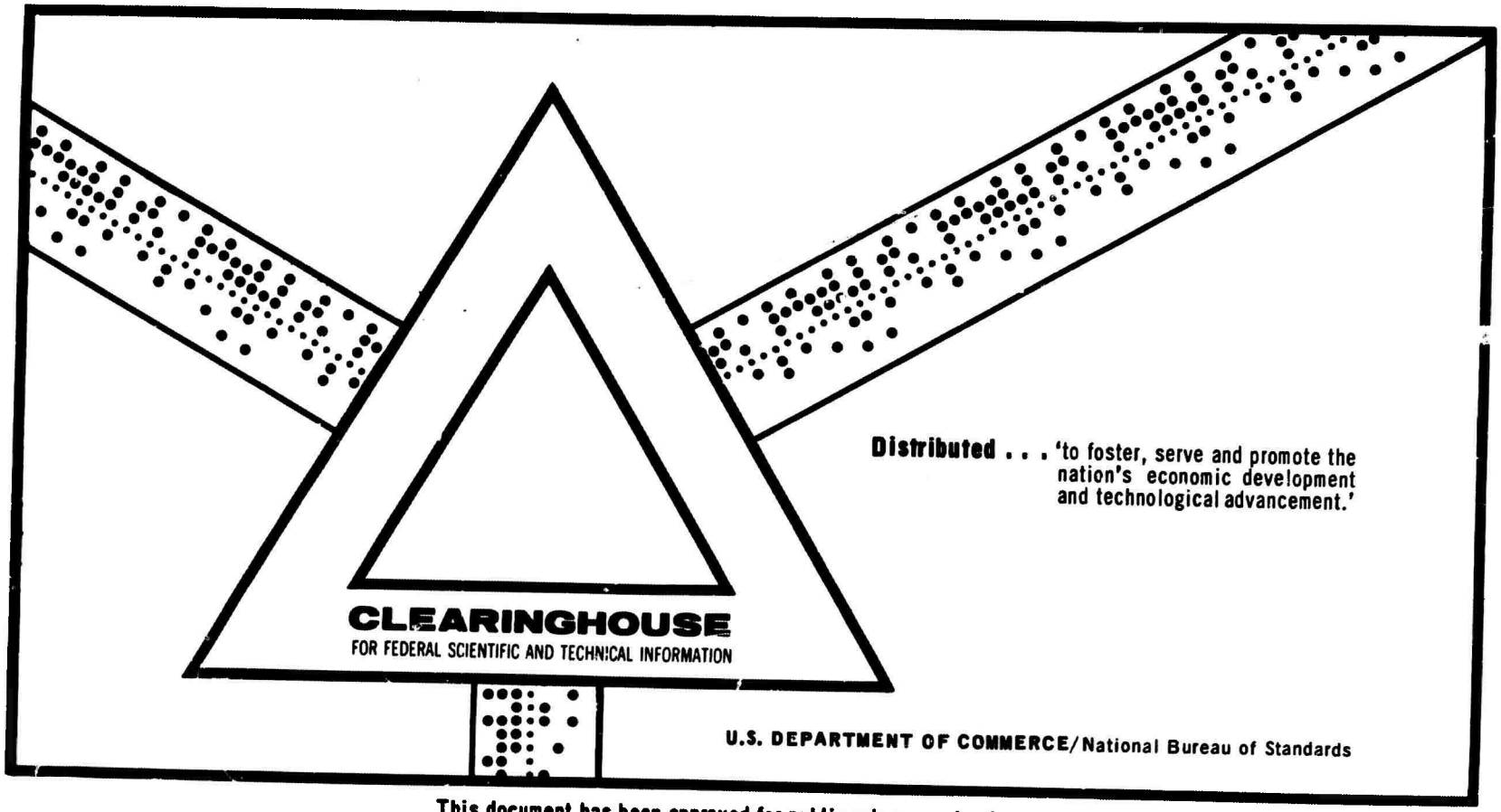
29 December 1969



December 29, 1969

Director
Advanced Research Projects Agency
Washington, D. C. 20301

Attention: Col. J. M. MacCall

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Short Title: Picosecond Laser Pulses
Subject: Quarterly Technical Report for the period 1 September 1969 to 30 November 1969
Reported by: M. E. Mack, E. B. Treacy, W. H. Glenn, A. J. DeMaria

Gentlemen:

Work during this period has been concerned with the continued investigation of stimulated scattering processes using picosecond pulse excitation, with an investigation of the behavior of the polarization of a pulse traveling in a Kerr active liquid and with an investigation of the generation of Cerenkov radiation by the nonlinear polarization induced in a dielectric by a high power optical pulse.

Stimulated Scattering

In the previous technical report, H-920479-17, Semi-Annual Report for the period 1 March 1969 to 31 August 1969, some experiments on stimulated transient Raman scattering in liquids were described. This work was performed by M. E. Mack of United Aircraft Research Laboratories and R. L. Carman, F. Shimizu and N. Bloembergen of Harvard University and has led to a paper entitled, "Forward Picosecond Stokes Pulse Generation in Transient Stimulated Raman Scattering" that has been accepted for publication in Physical Review Letters. This work has been extended to scattering in gases and the results are given in a report entitled, "Stimulated Rotational and Vibrational Raman Scattering in Gases". This report is

included as Appendix I to this report, and it is intended that this will be submitted for publication in Applied Physics Letters.

Nonlinear Propagation Effects

When an intense light wave propagates through a Kerr active liquid, the intensity dependent index of refraction can lead to a variety of effects such as self-focusing, self-phase modulation and steepening and alteration of the state of polarization of the wave. During this period, consideration has been given to the latter effect and several conclusions have been reached. It has been known for some time that when an elliptically polarized wave propagates through such a liquid, the orientation of the axis of the ellipse are rotated by an amount proportional to the product of the fields along the major and the minor axis and the distance traveled (Reference 1). This effect has been used to measure some of the nonlinear coefficients of the liquid (Reference 2). A somewhat more general approach to this effect has been carried out and it has been concluded that linear polarization is an unstable mode of propagation in such a liquid while circular polarization is a neutrally stable mode. The analysis has also been extended to the case where an additional linear birefringence, such as could be produced by an external electric field, is present. This investigation is still in progress; an outline of the method of approach and some of the main results is given below. A more detailed discussion will be reported upon completion of the analysis.

We consider the propagation of an optical pulse in a liquid having an anisotropic molecular polarizability. For the present, we will assume that the molecular reorientation time, τ, is short compared to the duration of the pulse. For typical liquids such as CS_2 and $C_6H_5NO_2$, τ is a few picoseconds to about 30 picoseconds. In this case we find that the polarization in the liquid can be written as

$$\bar{P} = \epsilon\bar{E} + \beta(I)\bar{E} + \bar{P}_a = \bar{P}_{LIN} + \bar{P}_{NL} \qquad \epsilon\bar{E} = \bar{P}_{LIN} \tag{1}$$

Here the first term represents the ordinary linear polarizability, the second term an intensity dependent but isotropic polarizability and the third term an anisotropic polarizability. We write the optical field as $R_e\ \bar{\mathcal{E}}e^{i(kz-wt)}$ and allow for the presence of a dc field E. The direction of propagation will be taken as the +z axis and the direction of the dc field will define the +x axis. The anisotropic part of the polarizability may then be expressed as

$$P_a = \alpha \begin{bmatrix} a & b \\ -b & 0 \end{bmatrix} E \tag{2}$$

with $a = E^2$ and $b = \frac{1}{4}(\mathcal{E}_y\mathcal{E}_x^* - \mathcal{E}_x\mathcal{E}_y^*)$. The fields may be calculated from the usual slowly varying envelope approximation,

$$\frac{\partial \mathcal{E}}{\partial Z} = i \frac{2\pi k}{\epsilon} P_{NL} e^{-ikz} \tag{3}$$

Substitution of Equation (1) and (2) into Equation (3) yields a set of two coupled equations in $\mathcal{E}_x$ and $\mathcal{E}_y$. It is convenient to write

$$\mathcal{E}_{x,y} = \mathcal{E}_{ox,oy} e^{i\phi_{x,y}(Z)} \tag{4}$$

Equating the real and imaginary parts of Equation (3) gives four equations for $\mathcal{E}_{ox}$, $\mathcal{E}_{oy}$, ϕ_x and ϕ_y. One integral follows immediately, i.e., $|\mathcal{E}_{ox}|^2 + |\mathcal{E}_{oy}|^2 =$ constant as would be expected since the material is lossless. We may then write

$$\begin{aligned} \mathcal{E}_{ox} &= I^{\frac{1}{2}} \cos\theta \\ \mathcal{E}_{oy} &= I^{\frac{1}{2}} \sin\theta \end{aligned} \tag{5}$$

Finally, solving for $\psi = \phi_y - \phi_x$ we find that the isotropic part of the nonlinear polarization drops out and we obtain two equations for θ and ψ.

$$\begin{aligned} \frac{d\theta}{dZ} &= \dot{\theta} = \gamma I \cos\theta \sin\theta \sin 2\psi \\ \frac{d\psi}{dZ} &= \dot{\psi} = \gamma_o + \gamma I(\cos 2\psi - 1)\cos 2\theta \end{aligned} \tag{6}$$

where $\gamma = \frac{\pi k \alpha}{4\epsilon}$, $\gamma_o = \frac{\pi k \alpha}{\epsilon} E^2$

These two equations describe completely the state of polarization of the wave as it propagates. The isotropic part of the nonlinear response does not enter because it does not change the state of polarization.

Equations(6) do not lead to a direct solution for $\theta(Z)$ and $\psi(Z)$however, it is possible to find a solution for ψ as a function of θ. We find

$$\sin^2\psi = \left[\frac{2\gamma_o}{\gamma I}(\cos 2\theta - \cos 2\theta_o) + \sin^2\psi_o \sin^2 2\theta_o\right]\left[\sin^2 2\theta\right]^{-1} \tag{7}$$

Here θ_o and ψ_o are constants that are determined by the initial conditions of polarization. This equation yields trajectories of ψ vs θ, the sense of traversal of these being determined by Equation (6). These are plotted schematically in Figure 1a for the case $\gamma_o = 0$ and Figure 1b for $\gamma_o \pm 0$. In all these trajectories, it should be

noted that a point sin ψ = +1, θ is identical to the point sin ψ = -1, $-\theta$. Thus, the curves in the upper right quadrant join continuously into those in the lower left and those in the upper left join those in the lower right. For the case γ_o = 0, the curves are the usual rotating polarization ellipse. The points sin $\psi = \pm 1$, $\theta = \pm \pi/4$ are the two senses of circular polarization. They are neutrally stabl., as they are surrounded by the trajectories. The case of linear polarization, ψ = 0, θ = arbitrary is seen to be unstable as all trajectories diverge from this line. The case $\gamma_o \neq 0$ corresponds to the presence of a linear birefringence. Here the trajectories split into two classes depending on the initial conditions and the intensity. One type is similar to the case γ_o = 0 and corresponds to a rotating (and slightly distorted) elliptical polarization. The other corresponds, for example, to an initial linear polarization at an angle θ. The polarization then becomes elliptical and rotates with initially increasing and then decreasing ellipticity until it becomes linear at $-\theta$. In the degenerate case I = 0, these are trajectories simply become θ = constant, $\psi = \gamma_o Z$ as may be seen directly from Equation (6). This is just the induced birefringence and the phase difference between the x and y components increases linearly with distance. The trajectories on a plot of sin ψ vs θ are two vertical lines at $\pm\theta$. This is physically identical to θ = constant ψ = arbitrary as noted following Equation (7).

It is anticipated that further work will be carried out in the analysis of these polarization effects. In particular, numerical or approximate analytical solutions for $\theta(Z)$ and $\psi(Z)$ will be obtained. It would also be of interest to extend this analysis to nonlinear materials with a more complicated spatial symmetry, i.e., nonlinear crystals such as calcite. It is expected that these nonlinear polarization effects will be important in the propagation of high power pulses in dielectric media, particularly in the polarization effects in self-trapped filaments.

Cerenkov Radiation from Picosecond Light Pulses

Cerenkov radiation is a very important phenomenon, which unfortunately has been largely neglected by physicists outside of the U.S.S.R. The main interest in this phenomenon in this country has centered around its use in relativistic charged particle detection. Following the original suggestion by Ginsberg (Reference 3) that Cerenkov radiation may be used as a source of microwave radiation, some efforts were made in this direction by various groups in this country. The most significant work along these lines was carried out by Professor Paul Coleman (Reference 4) and his associates at the University of Illinois.

The basic idea in the Cerenkov generation of microwaves is to shoot relativistic electron bunches through a narrow tunnel in a dielectric. The radiation comes out at wavelengths greater than the tunnel diameter. The Cerenkov cone geometry is governed by the ratio of the microwave phase velocity in the dielectric to the electron bunch velocity, which is approximately c for relativistic electrons. The radiated power from a compact bunch of N electrons is N^2 greater than that from a single electron. The basic experimental difficulty encountered is what Coleman calls the "beam-tunnel problem", which means the problem of generating and bunching relativistic electrons and shooting them down the narrow tunnel without having them hit the tunnel walls. A secondary problem was the dielectric losses at millimeter and submillimeter wavelengths.

We propose a more elegant approach to the Cerenkov generation problem based on nonlinear optical interactions. The basic Cerenkov radiation process is summarized as shown below:

Relativistic particle → Moving polarization → Radiation.

The alternative approach is:

Picosecond light pulse → Moving polarization → Radiation.

In the latter case, the polarization is produced by the "dc effect" of N.L.O. (rectification) in a transparent nonlinear crystal. The 1.06 micron wavelength of the light pulse is on the short wavelength side of the infrared resonances in the dielectric, so that its group velocity exceeds the phase velocities of all wavelength components belonging to the rectified pulse envelope. The rectified dipole moment per unit length replaces the charge of the electron bunch as the equivalent charge for wavelengths larger than the spatial dimensions of the pulse. The electron beam-tunnel problem is replaced by a free optical beam problem, which is obviously much simplier. Equivalent charges in excess of 10^8 electrons within submillimeter pulse dimensions over path lengths of a few centimeters would be trivial with our mode-locked laser and such common N.L.O. materials as KDP. The principal experimental difficulties are expected to be those of material damage.

The dielectric must now be anisotropic, which increases the difficulty of the theoretical calculations to some extent as compared to electrons in isotropic media. The theory that we are developing is capable of handling these complications as well as the complications of induced polarization direction.

The dc effect can produce transverse as well as longitudinal polarizations in the nonlinear medium, which adds another dimension to the Cerenkov problem, for even the Cerenkov radiation from an electric or magnetic dipole pointing transverse to the direction of its motion is not related to the radiation from a moving transverse polarization induced by a light pulse, so that the published results on the former cannot be used for the latter problem. The difference between the two cases is subtle. In brief, the moving transverse dipole gives rise to longitudinal equivalent field source currents in contrast to the transverse equivalent currents of the transverse N.L.O. rectified fields. A further distinction is apparent from the fact that the physical transverse dipole is subjected to the relativistic four-vector transformations that mix electric and magnetic moments.

All the classical Cerenkov problems relating to radiation from relativistic particles can be solved by linear operations on the field solutions for moving monopoles. Most of the relevant work has been done by Russian physicists, and the relevant English translations have proved to be difficult to acquire. As mentioned earlier, the scope of the N.L.O. Cerenkov interactions is considerably broader and much theoretical work needs to be carried out. The general problems to be solved are those involving the various Cerenkov field components (and hence radiated power, field polarizations, etc.) arising from spatially extended longitudinal and transverse

electric and magnetic polarizations in uniaxial and biaxial crystals where the polarization source is moving in an arbitary direction relative to the optic axis of the crystals, together with the coupling of the fields from the crystals to the surrounding space. Higher order effects involving Cerenkov radiation from certain moving entropy fronts deserve investigation. An example of this last mentioned effect would be the Cerenkov radiation from the nose of a trapped light filament in CS_2 in which the entropy change involves alignment of the CS_2 molecules along the field and the equivalent current sources arise from a term of the type $-(\partial/\partial t)$ div Q, where Q is the quadrupole moment per unit volume. Admittedly, the radiated power will be small, but the balance of field interactions that permit stable forward propagation of the nose of the trapped filament is delicate, and small interactions should not be arbitrarily dismissed.

A full and complete theoretical investigation of Cerenkov radiation in its most general ramifications will not be carried out under the present contract. A less ambitious theoretical program is under way, and when some theoretical results have been reduced to numbers, experimental demonstration of the effect will be planned. We feel that there are two important applications of Cerenkov radiation. One is the generation of broadband radiation bursts in the millimeter and submillimeter region of the spectrum. The other is as an intermediate step in the generation of subpicosecond pulses. We will be collaborating with Professor T. K. Gustafson of the Electrical Engineering Department at the University of California, Berkeley on this second application.

We now present a brief summary of our theoretical investigations to date. First, consider the possibility of using results that are already known from previous calculations on relativistic electrons. The classical problem is that of an electron with charge e_1 moving along the z-axis at constant speed v in a dielectric with refractive index m. The charge density is

$$\rho = e_1 \, \delta(x) \, \delta(y) \, \delta(z - vt) \tag{8}$$

which gives rise to a current density $\underset{\sim}{J}$ with the Fourier components

$$\underset{\sim}{J}_\omega = \hat{z} J_\omega = e_1 \hat{z} \, \delta(x) \, \delta(y) \, e^{i\omega z/v}/2\pi \tag{9}$$

The energy radiated per unit length per unit frequency interval is given by

$$\frac{d^2W}{d\omega d\ell} = \frac{e_1^2}{c^2} \left(1 - \frac{c^2}{v^2 m^2}\right) \omega. \tag{10}$$

For the isotropic medium under consideration, the solution is most easily obtained by solving the equation for the vector potential $\underset{\sim}{A}$:

$$(\nabla^2 + m^2\omega^2/c^2)\,\underset{\sim}{A}_\omega = \frac{-4\pi}{c}\,\underset{\sim}{J}_\omega \tag{11}$$

where the current density $\underset{\sim}{J}$ is given by Equation (2). Now suppose that by some method a longitudinal polarization pulse is made to travel along the z-axis of the isotropic dielectric at speed v. Its radiation can be calculated immediately from the results above. To be more specific, consider a longitudinal polarization front of width Λ limited to the immediate neighborhood of the z-axis:

$$\underset{\sim}{P} = \hat{z}Q\left[\tfrac{1}{2} - \frac{1}{\pi}\tan^{-1}\{(z - vt)/\Lambda\}\right]\delta(x)\,\delta(y) \tag{12}$$

so that

$$\underset{\sim}{J}_\omega = -\hat{z}(2\pi)^{-1}\,Qe^{-\Lambda|\omega|/v}\,e^{i\omega z/v}\,\delta(x)\,\delta(y) \tag{13}$$

Comparing Equation (13) with Equation (9), we see that we should replace e_1^2 in Equation (10) by $\left[Qe^{-\Lambda|\omega|/v}\right]^2$. For a source extended in the x and y directions, two additional integrations (over x and y) will be needed in calculating the field components, and this step is straightforward.

The above example is unrealistic in that we considered an isotropic medium. However, the same technique of Fourier decomposition can be used to compute the radiation in optically anisotropic media caused by longitudinal polarization simply by substitution into the known results for relativistic particles in optically anisotropic media. Thus half the problems for Cerenkov radiation from picosecond pulses are solved automatically. The other half are those involving transverse nonlinear polarizations.

To get a ready comparison between the radiations from longitudinal and transverse polarizations, it again is easiest to refer back to the hypothetical case of an isotropic medium. In this case, the inhomogeneous term in Equation (11) becomes $4\pi i(\omega/c)\,\hat{x}\,f(\rho)\,\delta(z - vt)$ where $f(\rho)$ indicates the transverse extent of the cylindrically symmetric polarization and the δ-function in z is not restrictive but can be used as the basic element for a later integration over z. With the substitution

$$\underset{\sim}{A}_\omega = u\,e^{i\omega z/v}(\hat{\rho}\cos\phi - \hat{\phi}\sin\phi) \tag{14}$$

it is easy to shown that u satisfies a Bessel equation and is given by

$$u(\rho) = \frac{4\pi^2\omega}{vc}\,H_1^{(1)}(s\rho)\int_0^\infty f(\rho_o)J_1(s\rho_o)d\rho_o \tag{15}$$

with $s = \frac{m\omega}{c}\left(1 - \frac{c^2}{m^2v^2}\right)^{\frac{1}{2}}$.

The energy radiated per unit length per unit frequency interval is

$$\frac{d^2W}{d\ell\, d\omega} = \frac{\omega\pi}{4}\left(1 + \frac{c^2}{m^2v^2}\right) i\rho\, \left(u\frac{d\bar{u}}{d\rho} - \bar{u}\frac{du}{d\rho}\right) \tag{16}$$

If $f(\rho_o)$ equals the constant value a for $\rho_o \leq R$ and zero elsewhere, where sR is small for all s of interest the energy formula becomes

$$\frac{d^2W}{d\ell\, d\omega} = \frac{4}{9}\pi a^2(\pi R^2)^3 \frac{m^2\omega^5}{v^2c^4}\left[1 - \left(\frac{c^2}{m^2v^2}\right)^2\right] \tag{17}$$

This energy is small compared to that for longitudinal polarization, the smallness being due to the fact that the radiation field goes as the Hankel function $H_1(s\rho)$ so that the source strength is measured by its overlap on the first order Bessel function $J_1(s\rho)$ as in Equation (15). Clearly, one should work with longitudinal polarizations if possible.

Considering the radiation from a longitudinal polarization in an optically anisotropic medium as known by correspondence with the charged particle results, there remains the problem of solving for the radiation from a transverse polarization. This problem has been formulated, again by a Fourier decomposition technique. For motion along the z-axis, the polarization is resolved into a continuous angular spectrum of plane waves where the z and t dependences are exp $i\omega(z/v - t)$. For each plane wave of polarization, the electromagnetic field components in the anisotropic medium can be found by the method described by Kleinman (Reference 5). This involves the complication that the electric field for one plane wave component is resolved into skew components, the skew axis directions being continuously variable in the plane wave spectrum variables. In brief, a Fourier component of the polarization field couples strongly to an electromagnetic wave component if their phase velocities match, with the result that both ordinary and extraordinary cones of Cerenkov radiation are emitted. This part of the calculation is almost complete for the case of uniaxial crystal with the transverse polarization source moving along the optic axis.

During the next reporting period, research will continue in the areas discussed above.

Very truly yours,

William H. Glenn

William H. Glenn
Principal Scientist
Quantum Physics

WHG/der

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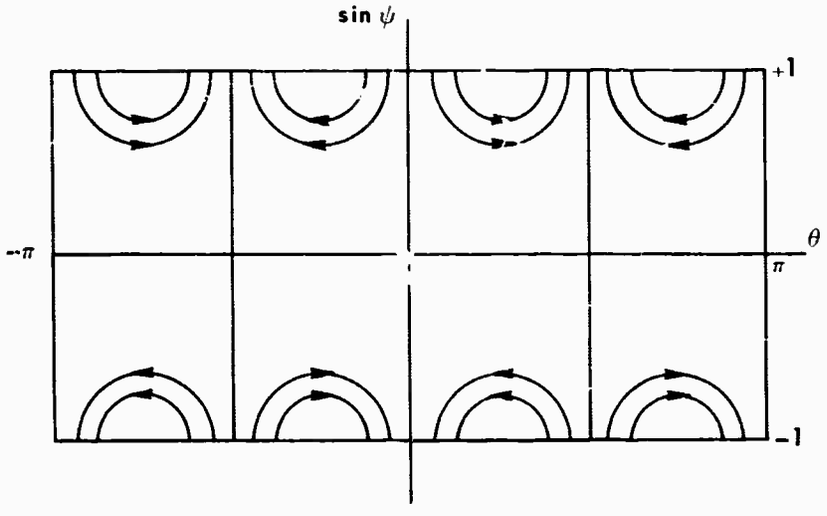
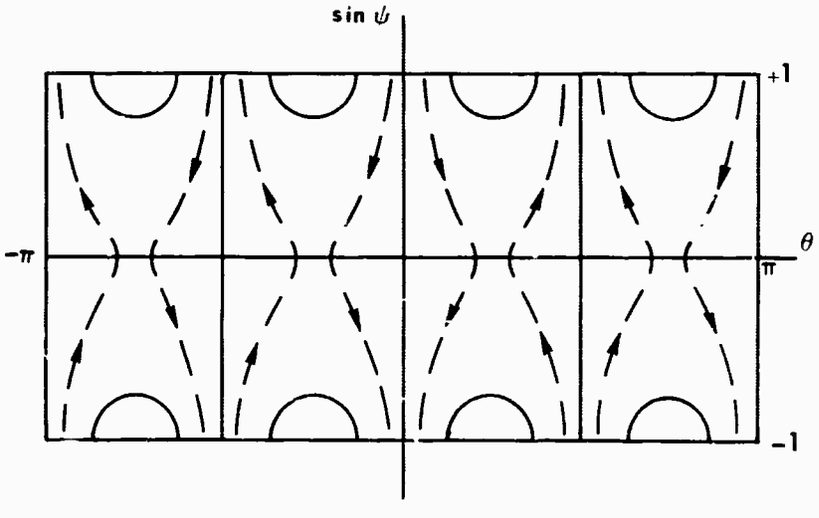
FIG. 1

BEHAVIOR OF POLARIZATION



KERR ACTIVE LIQUID



KERR ACTIVE LIQUID
WITH INDUCED BIREFRINGENCE

APPENDIX I

Transient Stimulated Rotational and Vibrational Raman Scattering in Gases

Paper prepared for submission to Applied Physics Letters.

H920479-19

TRANSIENT STIMULATED ROTATIONAL AND VIBRATIONAL RAMAN SCATTERING IN GASES

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ABSTRACT

Using a mode-locked ruby laser, stimulated rotational and vibrational Raman scattering has been observed in a variety of high pressure gases, where previously no stimulated scattering has been reported. Pulse energy conversion efficiencies as high as 70% have been obtained. Evidence for a strong optical Stark effect is seen in the gases producing rotational scattering. Self focussing in a collimated beam is observed in N_2O and CO_2.

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Considerable experimental effort has been spent on stimulated Raman scattering in high pressure gases, much of it motivated by the possibility of obtaining new high brightness lasers. Only methane, hydrogen and deuterium have produced stimulated Raman scattering with the Q-switched lasers used[1-3]. Rotational scattering has been observed in hydrogen and in deuterium[4]. In this paper we wish to report the observation of stimulated vibrational scattering in a variety of gases using a mode-locked ruby laser. In addition, stimulated pure rotational and rotational-vibrational Raman scattering has been observed in oxygen, carbon dioxide and nitrous oxide.

Recent experiments with picosecond laser pulses have probed stimulated Raman scattering in liquids under transient conditions[5-10]. Theoretical studies[11,12] indicate that typical transient effects should occur, if the duration τ_p of the exciting pulses is reduced below $G_{ss}\tau_\nu$, where G_{ss} is the steady state gain coefficient and τ_ν is the molecular vibrational or rotational dephasing time. For $t_p \ll G_{ss}\tau_\nu$, the Raman gain will decrease, the Stokes pulses will narrow in time

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rather than in frequency, and the transient gain coefficient will be proportional to the total integrated cross section of the Raman line and independent of its width $1/\tau_\nu$. Experiments in liquids have verified these novel features of transient stimulated Raman scattering[10]. As a result of the transiency it is possible to excite broad Raman lines with picosecond pulses[8-10], which cannot reach threshold in the steady state regime. Transient excitation with picosecond laser pulses can be also used to advantage to discriminate against more slowly responding competing gain processes such as Brillouin scattering. This is expecially valuable in gases, since with Q-switched pulses Brillouin scattering is often the dominant nonlinearity.[2]

To observe stimulated scattering, a test cell 54 cm. long was pressurized with the sample gas. The output of a mode-locked ruby laser[13], producing a train of 5 psec. duration pulses with a maximum peak power of about 5 Gw. in a 0.5 $cm.^2$ beam, was then focussed into the cell with a 50cm. focal length lens. The spectra of the stimulated scattering from gases in the cell were recorded on Kodak IN plates using a Jarrell-Ash 75-152 Spectrograph. The experimental results for stimulated vibrational Raman scattering are summarized in Table I.

The largest conversion efficiencies occurred for CH_4 and SF_6. In both cases, more than twenty percent of the total incident laser energy was converted to Stokes energy. In CH_4 stimulated scattering could be produced without focussing. Stimulated vibrational scattering in a collimated beam could also be generated in SF_6, N_2 and CO_2 by telescoping down the incident ruby laser beam with a 2 power Galilean telescope. Nearly all of the conversion takes place in the early pulses of the train, as is shown in Fig. 1(a) and 1(b) for SF_6. Up to seventy percent of the energy in these initial pulses could be converted to Stokes energy as verified by

the depletion of the incident laser in figure 1(a) and by direct energy measurements. The reason for the decrease in conversion towards the end of the train is not clear, but it may represent a saturation of the vibrational population. Another possibility is the build up of slower thermal or acoustic perturbations such as discussed by Pohl[14], and observed recently by one of the present authors[15].

In the case of CO_2, O_2 and N_2O, stimulated pure rotational and rotational-vibrational Raman scattering could also be produced if the incident ruby beam is circularly polarized[4]. In N_2O, which has the largest rotational cross section of the three gases, both first and second rotational and rotational-vibrational Stokes lines are excited. Evidence of rotational scattering is also seen in propylene. Figure 2 shows pure rotational Raman spectra and corresponding desitometer traces for CO_2 and O_2. In CO_2 the lines S(12) through S(24) have been stimulated, while in O_2, S(5) through S(13) have been stimulated. The line shifts observed agree within experimental error with those reported for the spontaneous spectra[16]. In the case of N_2O, the narrowness of the line spacing in comparison to the linewidths precludes an accurated identification of the stimulated lines. The observed rotational scattering spans the region from S(13) to S(20).

Often the rotational lines observed are broadened and skewed from line center or even split into several components. This is believed to be the result of an optical Stark effect. Neglecting any permanent molecular dipole moments, the Stark shift for circularly polarized light is to lowest order in perturbation theory

$$\Delta W = \frac{4\pi}{3}\frac{\gamma I}{c}\frac{J(J+1) - 3m^2}{(2J-1)(2J+3)} \qquad (1)$$

where the z-axis has been chosen in the direction of propagation. Here γ is the anisotropic part of the polarizibility tensor and I is the incident intensity.

Using (1), one may readily confirm that the shifts are an appreciable fraction of the total rotational energy for the J-levels involved, and that the splitting in the rotational shifts for the different m-states is of the order of a few reciprocal centimeters, in qualitative agreement with the splitting observed. In gases, where rotational freedom is preserved, the Stark shifts are also responsible for the observed Kerr effect. For $\gamma>0$ in Eq. (1), the lowest energy states are $m= \pm J$, corresponding to molecular rotation in the plane of the electric field. This is equivalent to classical alignment. Further evidence of the importance of the optical Stark effect in the transient regime is provided by the observation of self trapping in a collimated beam in the most anisotropic gases, CO_2 and N_2O . This is most clearly seen in vibrational Stokes near field patterns such as shown in figure 1(c) for CO_2.

If linearly polarized light is used in an attempt to observe stimulated rotational Raman scattering, one sees instead a broad wing shaded to the Stokes side on both the ruby line and on the vibration Stokes line. Similar results are found in the case of Rayleigh wing scattering[17]. Explanations for the difference in behavior with circular and linearly polarized light have been given by Herman[18] and by Chiao and Godine[19]. The sense of circular polarization of the stimulated first rotational and rotational-vibrational lines is always reversed from that of the incident laser light. The same situation is found in the stimulated rotational scattering from hydrogen[4] and in Rayleigh wing scattering[17]. These experimental results are in agreement with theoretical predictions giving a forward gain ratio for the opposite to same sense of polarization of 6:1, if the anti-stokes coupling is ignored[4] and 6:0, if this coupling is taken into account[19]. The second rotational and rotational-vibrational Stokes lines observed in N_2O are polarized

in the same sense as the laser light, and, therefore, in the opposite sense as the first rotational Stokes lines, as expected from the sequential nature of the higher order Stokes production.

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FIGURE CAPTIONS

Figure 1. Vibrational Scattering

a. Interleafed Incident and Transmitted Laser Pulse Trains Showing Depletion Due to Raman Scattering in SF_6. (Incident Train Slightly Delayed.)

b. Simultaneous Stokes Oscillograph.

c. Stokes Emission in CO_2 from Self Trapped Filaments.

Figure 2. Pure Rotational Raman Scattering

a. Spectrum with CO_2 at 30 psig.

b. Densitometer Tracing Indicating Initial J States.

c. Spectrum with O_2 at 900 psig.

d. Densitometer Tracing.

Table I - Vibration Raman Lines Stimulated

	GAS	PRESSURE (PSIG)	OBSERVED STIMULATED SHIFTS (cm^{-1})	CORRESPONDING SPONTANEOUS SHIFTS (cm^{-1})	DESIGNATION
N_2	Nitrogen	800-1500	2330	2330.7	ν_0
O_2	Oxygen	700-1500	1550	1554.7	ν_0
CO_2	Carbon Dioxide	250- 750	1385	1388.3.	ν_1
N_2O	Nitrous Oxide	700	1282	1285.0	ν_1
			774	775	ν_1
SF_6	Sulfur Hexafluoride	200- 280	1551	1550	$2\nu_1$
			2323	2325	$3\nu_1$
CH_4	Methane	1200-1400	29.6	2914.2	ν_1
C_2H_4	Ethylene	800	1344	1342.4	ν_3
C_3H_6	Propylene	1300	~2920	2924(liq.)	ν_4
HCl	Hydrogen Chloride	480	2883	2886.0	ν_0
HBr	Hydrogen Bromide	300	2558	2558	ν_0

H920479-19

FIG. 1

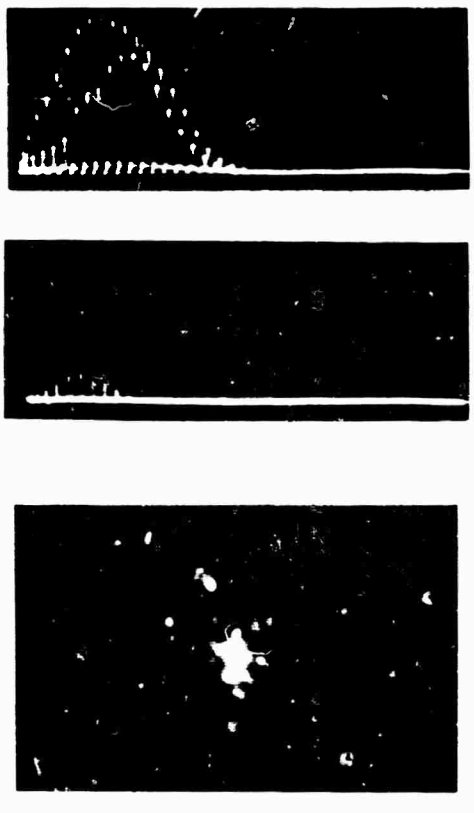

FIG. 2

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13. ABSTRACT

~~This~~ report describes experimental investigations of stimulated rotational and vibrational Raman scattering in gases, analytical investigations of nonlinear propagation effects in Kerr active liquids and a discussion of the generation of Cerenkov radiation by light pulses.

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